

Superconducting gap structure of CeIrIn₅ from field-angle-resolved measurements of its specific heat

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In order to identify the gap structure of CeIrIn₅, we measured field-angle-resolved specific heat $C(\phi)$ by conically rotating the magnetic field H around the c axis at low temperatures down to 80 mK. We revealed that $C(\phi)$ exhibits a fourfold angular oscillation, whose amplitude decreases monotonically by tilting H out of the ab plane. Detailed microscopic calculations based on the quasiclassical Eilenberger equation confirm that the observed features are uniquely explained by assuming the $d_{x^2-y^2}$ -wave gap. These results strongly indicate that CeIrIn₅ is a $d_{x^2-y^2}$ -wave superconductor and suggest the universal pairing mechanism in $CeMIn_5$ ($M = Co, Rh$, and Ir).

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The heavy-fermion systems $CeMIn_5$ ($M = Co, Rh$, and Ir) have been extensively studied because they exhibit unconventional superconductivity near the antiferromagnetic (AF) quantum critical point (QCP). Especially, much effort has been expended to identify the superconducting (SC) gap structure, a challenging issue that is closely related to the identification of the pairing mechanism. Recently, field-angle-resolved experiments along with theoretical works have confirmed that CeCoIn₅ is a $d_{x^2-y^2}$ -wave superconductor.¹⁻³ Because of the $d_{x^2-y^2}$ -wave gap and the proximity of the system to the AF state, it is now widely accepted that the pairing interaction in CeCoIn₅ is AF spin fluctuations. Pressure induced superconductivity in CeRhIn₅ has also been examined by field-angle-resolved specific heat measurements and it is argued to have the same pairing symmetry.⁴

By contrast, the possibility of a different pairing mechanism has been suggested for CeIrIn₅ due to its unusual behavior of the SC phase.⁵ The transition temperature T_c , which is 0.4 K at $P=0$, increases to 0.8 K under a high pressure of $P=2.1$ GPa,⁶ although strong AF fluctuations existing at ambient pressure are rapidly suppressed by increasing P as the system is further pushed away from a hypothetical AF QCP. Moreover, when Rh is doped to Ir sites, T_c shows a cusp-like minimum before reaching the maximum T_c of ~ 1 K around the onset of an AF state.^{7,8} These observations suggest the presence of two distinct SC phases in CeRh_{1-x}Ir_xIn₅.

While several attempts have been made to uncover the gap structure of CeIrIn₅, two conflicting possibilities have remained. Kasahara *et al.*⁹ reported a fourfold angular oscillation in the thermal conductivity $\kappa(\phi)$ when H is rotated in the basal plane, and attributed its origin to the vertical line nodes of the $d_{x^2-y^2}$ -wave gap. On the other hand, Shakeripour *et al.*^{10,11} examined the effect of impurity scattering of $\kappa(T)$ for a current parallel and perpendicular to the c axis, and proposed the gap function of CeIrIn₅ to be either k_z or $k_z(k_x + ik_y)$, both of which have a horizontal line node only on the equator, in sharp contrast to the $d_{x^2-y^2}$ -wave gap. At present, neither of these two possibilities can be ruled out. The temperature variation of the anisotropy κ_c/κ_a in Ref. 10 can be explained

within the $d_{x^2-y^2}$ symmetry if the Fermi surface has small deviations from the cylindrical symmetry,¹² whereas the fourfold oscillation in $\kappa(\phi)$ can be explained by the horizontal line node gap if an in-plane anisotropy of the effective mass and/or minima exist in the azimuthal variation of the gap amplitude.¹¹ Thus, the gap structure of CeIrIn₅ has remained controversial.

In order to settle the controversy over the gap structure of CeIrIn₅, we have performed field-angle-resolved specific heat $C(\phi, \theta)$ measurements down to 80 mK. The $C(\phi)$ measurements, where ϕ denotes the in-plane azimuthal angle of H , have proven to be quite useful to determine the direction of nodes in the momentum space of bulk superconductors.¹³ Here we extend the method to measure the polar angle θ dependence of $C(\phi)$ by which a detection of the horizontal line node can be made. We revealed that $C(\phi, \theta)$ exhibits a clear fourfold oscillation as a function of ϕ with H rotated around the c axis, and its amplitude is monotonically suppressed by tilting H out of the ab plane (decreasing θ from 90°). The results are compared with the microscopic theory by solving the quasiclassical Eilenberger equation self-consistently, and are found to be in good agreement with the $d_{x^2-y^2}$ -wave gap, but are in a sharp contrast with the behavior predicted for the horizontal line node gap.

The single crystal of CeIrIn₅ used in the present study ($T_c = 0.4$ K, 40.4 mg) was grown by the self-flux method. The specific heat was measured by the relaxation and the standard adiabatic heat-pulse methods in a dilution refrigerator (Oxford Kelvinox AST Minisorb). Magnetic fields were applied in the xz plane by using a vector magnet consisting of horizontal split-pair (5 T) and vertical solenoid (3 T) coils. By rotating the refrigerator around the z axis using a stepper motor mounted at the top of a magnet Dewar, three dimensional control of the magnetic field direction is achieved. We confirmed that the addenda contribution was always less than 5% of the sample specific heat and had no field-angle dependence. An accurate ($< 0.1^\circ$) and precise ($< 0.01^\circ$) field alignment with respect to the crystalline ab plane was accomplished by making use of the $C(\theta)$ data which reflects the tetragonal anisotropy of H_{c2} .

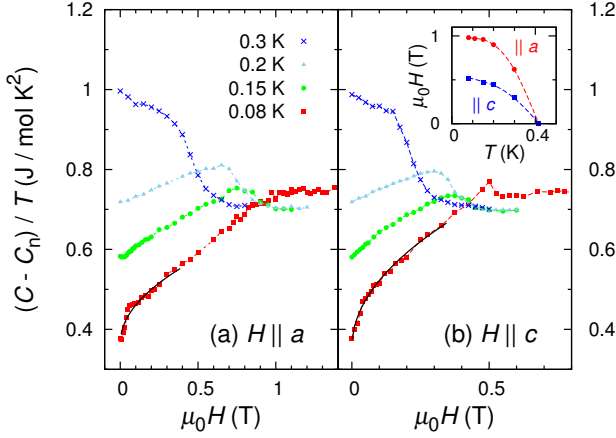


FIG. 1: (Color online) Magnetic field dependence of the nuclear subtracted specific heat divided by temperature C_e/T for (a) $H \parallel [100]$ and (b) $H \parallel [001]$. The solid lines represent fits to the data by $a\sqrt{H} + b$. The inset in (b) shows the temperature dependence of the upper critical field determined from the present study.

The specific heat of CeIrIn_5 at zero field is known to exhibit an upturn on cooling below about 0.1 K due to a quadrupole splitting of the ^{115}In ($I = 9/2$) and $^{191,193}\text{Ir}$ ($I = 3/2$) nuclear spins.^{14,15} In this paper, the nuclear Schottky contribution was subtracted from the data assuming $C_n = (a_0 + a_1 H^2)/T^2$, where a_0 was adjusted so that the resulting electronic contribution $C_e = (C - C_n)$ at $H = 0$ becomes proportional to T^2 at low T . The value of a_0 ($= 0.38$ mJ K/mol) thus obtained was in good agreement with the calculated one (~ 0.34 mJ K/mol) using the parameters determined from the ^{115}In nuclear quadrupole resonance experiment.^{16,17} The coefficient a_1 was calculated from the nuclear Zeeman splitting. It can be shown that C_n is independent of the in-plane field orientation.¹ Because of the nuclear Schottky contribution, the $C(\phi, \theta)$ measurements were limited to the temperature range $T \geq 80$ mK.

Figures 1(a) and 1(b) show $C_e(H)$ for $H \parallel a$ and $H \parallel c$, respectively. As represented by the solid lines, $C_e(H)$ is proportional to the square root of H at low H and low T . This behavior supports the presence of line nodes in the SC gap. The $C_e(H)$ behavior near H_{c2} is in good agreement with the calculated result for a two-dimensional d -wave superconductor with a relatively small Pauli paramagnetic parameter $\mu = 0.86$.¹⁸ At 80 mK for $H \parallel a$, a cusp-like structure is observed at 0.06 T, which might originate from the multi-gap superconductivity, as reported in Sr_2RuO_4 (Ref. 19) and MgB_2 .²⁰

Based on the $C_e(H)$ data, we determined the upper critical field $H_{c2}(T)$, as plotted in the inset of Fig. 1(b). At the lowest temperature 80 mK, H_{c2} of the present sample is 1.0 T for $H \parallel a$ and 0.55 T for $H \parallel c$. The reduced ratio $\alpha = -H_{c2}/(T_c dH_{c2}/dT|_{T=T_c})$ is estimated to be 0.47 for $H \parallel a$ and 0.54 for $H \parallel c$, which agree well with the results of Ref. 21. These results also suggest that the Pauli paramagnetic effect in CeIrIn_5 is relatively weak compared with that in CeCoIn_5 [$\alpha = 0.26$ (Ref. 21)].

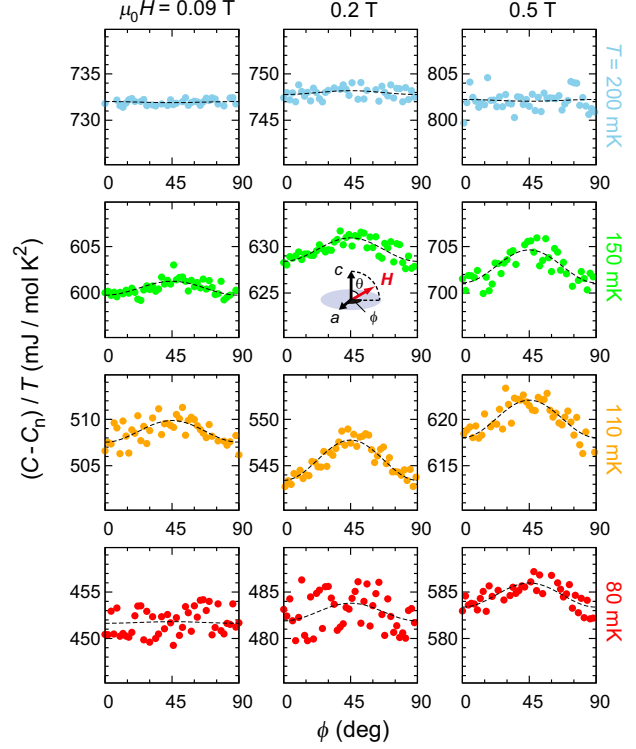


FIG. 2: (Color online) Variations of C_e/T as a function of the azimuthal angle ϕ between the $[100]$ axis and the magnetic field applied in the ab plane ($\theta = 90^\circ$). The dashed lines represent fits to the data by $C_0(T) + C_H(T, H)(1 - A_4 \cos 4\phi)$.

Figure 2 shows the ϕ dependence of C_e/T obtained by rotating H in the ab plane, where ϕ is measured from the $[100]$ direction. For each data point, the $C_e(\phi)$ value was determined by an average of ten successive measurements. The error bar is estimated to be several mJ/mol-K² for $T \geq 110$ mK, while it becomes about 10 mJ/mol-K² at 80 mK due to the large nuclear contribution. A clear fourfold oscillation was observed in $C_e(\phi)$ in the wide T and H region. We carefully confirmed the absence of a twofold oscillation in $C_e(\phi)$ which guarantees the accurate and precise alignment of the magnetic field with respect to the ab plane. In this intermediate T regime ($0.2 \leq T/T_c \leq 0.5$), $C_e(\phi)$ becomes minimum in fields along the $\langle 100 \rangle$ directions.

To characterize the H and T variations of the fourfold oscillation, we fit the data to the expression

$$C_e(T, H, \phi) = C_0(T) + C_H(T, H)(1 - A_4 \cos 4\phi) \quad (1)$$

as represented in Fig. 2 by the dashed lines. Here, C_0 and C_H are the zero-field and field-dependent components of C_e , respectively, and A_4 is the amplitude of the fourfold oscillation normalized by C_H . Figure 3(a) shows the T dependence of A_4 at $\mu_0 H = 0.09, 0.2$, and 0.5 T. We found that $A_4(T, H)$ exhibits a peak at around $0.3T_c$ and $0.15H_{c2}$, and rapidly decreases down to zero around $0.2T_c$. This is a feature that, to the best of our knowledge, has not been observed in previous $\kappa(\phi)$ measurements done mainly above $0.4T_c$,⁹ and strongly suggests the

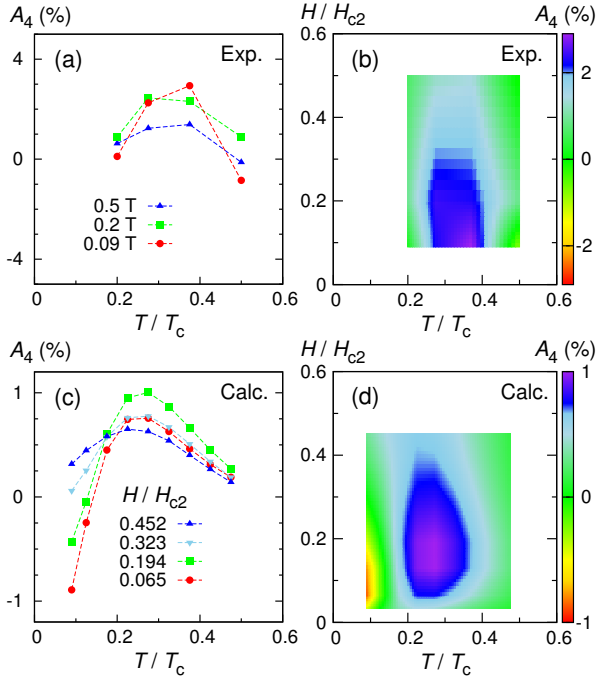


FIG. 3: (Color online) Temperature dependence of the normalized fourfold amplitude A_4 at several fields obtained by (a) the present experiment and (c) the microscopical calculations assuming the $d_{x^2-y^2}$ -wave gap.²² (b), (d) Contour plots of $A_4(T, H)$ using the same data in (a) and (c), respectively. Here, $T_c = 0.4$ K and $\mu_0 H_{c2} = 1$ T.

existence of fourfold vertical line nodes. These features can be seen more clearly by the contour plot of $A_4(T, H)$ in Fig. 3(b).

The ϕ rotation experiment alone, however, cannot rule out the possibility of the horizontal line node as claimed in Ref. 11. In order to solve this issue, we investigated $C_e(\phi)$ by conically rotating H at several fixed θ , where θ denotes the polar angle between H and the c axis. The results obtained at 110 mK are shown in Fig. 4(a), where the dashed lines are the fit to Eq. (1). In these measurements, the intensity of H is adjusted for each θ to satisfy $H/H_{c2}(\theta) = 0.2$ in order to avoid unfavorable effects of the tetragonal anisotropy in H_{c2} . The θ dependence of H_{c2} was experimentally determined from the $C_e(H)$ measurements at $T=110$ mK and is shown in Fig. 4(b) by the solid circles. The solid line in the figure is a fit to the Ginzburg-Landau anisotropic-effective-mass formula for three-dimensional superconductors²³ $H_{c2}(\theta) = H_{c2||c}/(\cos^2 \theta + (H_{c2||a}/H_{c2||c})^2 \sin^2 \theta)^{1/2}$, which reproduces the observed θ variation of H_{c2} satisfactorily. Figure 4(c) shows the θ dependence of A_4 obtained from the results in Fig. 4(a). We found that A_4 decreases gradually and monotonically with decreasing θ from 90° ($H \perp c$) to 0° .

Let us compare the experimental results with microscopic calculations and discuss the SC gap structure of CeIrIn₅. We assume a two-dimensional cylindrical Fermi surface for the main Fermi surface of CeIrIn₅ with significant f -electron contribution.²⁴ The local density of states (LDOS) of the quasiparticles (QPs) under the rotating H was calculated by

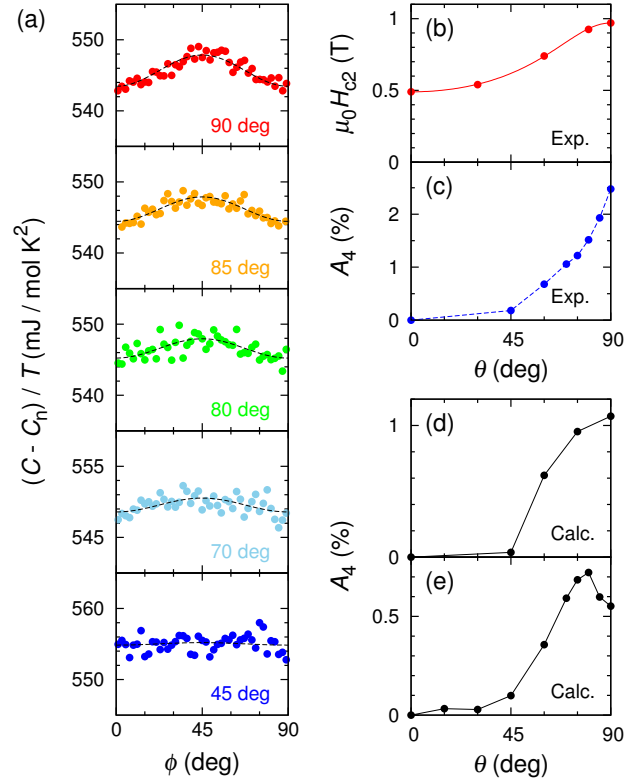


FIG. 4: (Color online) (a) Field-angle ϕ dependence of C_e/T in a conically rotating magnetic field with a strength of $0.2H_{c2}(\theta)$ at several fixed polar angles θ , measured at 110 mK. The dashed lines are fits to the data by the expression $C_0(T) + C_H(T, H)(1 - A_4 \cos 4\phi)$. (b) and (c) show the polar angle θ dependence of H_{c2} and A_4 at 110 mK, respectively. The calculated θ dependence of the zero-energy DOS anisotropy $[N_0(H \parallel \text{antinode}) - N_0(H \parallel \text{node})]/2N_0(H)$ is shown in (d) and (e) for the order parameters $k_x^2 - k_y^2$ and $k_z(k_x^2 - k_y^2)$, respectively.

solving the quasi-classical Eilenberger equation self consistently. Here, we assume the gap function to be $k_x^2 - k_y^2$, and no Pauli paramagnetic effect ($\mu = 0$) is considered. The H and T variations of the A_4 coefficient were then evaluated from the LDOS²² and the results are shown in Figs. 3(c) and 3(d) (contour plot). In the low H ($< 0.2H_{c2}$) and low T ($< 0.12T_c$) region, A_4 becomes negative implying that the $C_e(\phi)$ oscillation has minima along the nodal directions ($\langle 110 \rangle$). In this regime, A_4 is approximately proportional to the anisotropy of the zero-energy DOS N_0 [$A_4 \propto 1 - N_0(H \parallel \text{node})/N_0(H \parallel \text{antinode})$]²⁵ that can be intuitively understood by the Doppler-shift effect of the QPs on the circulating supercurrent around the vortices. The Doppler shift is given by $\delta = m_e \mathbf{v}_F \cdot \mathbf{v}_s$, where m_e is the electron mass, \mathbf{v}_F is the velocity of the QP and \mathbf{v}_s is the local superfluid velocity that is perpendicular to H . For a superconductor with line nodes, N_0 is enhanced by the Doppler shift of the nodal QPs, giving rise to the $H^{1/2}$ behavior of $C_e(H)$. If H is in a nodal direction, then those QPs cannot contribute to N_0 because δ vanishes. As a consequence, N_0 exhibits an angular oscillation with $N_0(H \parallel \text{node}) < N_0(H \parallel \text{antinode})$.

As T increases, contributions of the finite-energy DOS to A_4 become relevant.^{3,26} The detailed calculations tell us that the sign of A_4 changes around $0.15T_c$ as shown in Fig. 3(c), and $C_e(\phi)$ takes minima for $H \parallel$ antinodal directions in the intermediate T range. These theoretical results well reproduce our experimental data as can be seen by comparing the contour plots in Figs. 3(b) and 3(d). Especially, the peak position and the line of the sign change in $A_4(T, H)$ agree remarkably with each other. The agreement becomes worse if a relatively strong Pauli effect is introduced ($\mu=2$ – see Fig. 11 in Ref. 22). This fact is compatible with the weak Pauli effect ($\mu < 1$) expected from $C_e(H)$. Although the low- T Doppler-shift-predominant region could not be reached in our experiment, our data strongly indicate nodes along $\langle 110 \rangle$ directions.

In order to provide further evidence of the $d_{x^2-y^2}$ -wave gap in CeIrIn₅, we calculated the θ dependence of A_4 . Here we compare two types of gap functions $k_x^2 - k_y^2$ and $(k_x^2 - k_y^2)k_z$, and the results are shown in Figs. 4(d) and 4(e). The latter mimics the case of a horizontal line node gap with an azimuthal angular variation of the gap amplitude as claimed in Ref. 11. For the $d_{x^2-y^2}$ -wave gap, a gradual and monotonic decrease of A_4 on decreasing θ from 90° is expected [Fig. 4(d)], whereas for the $(k_x^2 - k_y^2)k_z$ gap $A_4(\theta)$ is predicted to have a dip around $\theta = 90^\circ$ [Fig. 4(e)]. The reason why $A_4(\theta)$ shows a local minimum at $\theta = 90^\circ$ for the horizontal line node gap is because there always exist nodal QPs whose momentum is parallel to the field direction, irrespective of ϕ . In this case, the anisotropy in the Doppler shift δ is strongly reduced, leading to the reduction of A_4 . It is obvious that the $k_x^2 - k_y^2$ gap

rather than the horizontal line node gap better reproduces the experimental data in Fig. 4 (c). In particular, the absence of the dip of $A_4(\theta)$ at $\theta = 90^\circ$ in our results strongly indicates that CeIrIn₅ does not have a horizontal line node at $k_z = 0$. These observations lead us to conclude that the SC gap in CeIrIn₅ is of $d_{x^2-y^2}$ type.

In conclusion, we have performed field-angle-resolved specific-heat measurements on CeIrIn₅ with $T_c = 0.4$ K by conically rotating the magnetic field around the c axis. The specific heat exhibits a clear fourfold angular oscillation and its dependences on temperature and field is surprisingly well reproduced by the microscopic calculations for a $d_{x^2-y^2}$ -wave superconductor. One of the most important findings in the present study is the monotonic variation of the oscillation amplitude with the polar angle of a conically rotating field. This feature confirms the absence of a horizontal line node on the equator and strongly supports the $d_{x^2-y^2}$ -wave gap, as in CeCoIn₅ and CeRhIn₅. The establishment of the identical gap symmetry in Ce M In₅ ($M = \text{Co, Rh, and Ir}$) indicates the universality of the pairing mechanism in this family and provides important hints for resolving the mechanism of the unique superconductivity.

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Note added in proof.— Recently, Lu *et al.* also reported $C(\phi)$ of CeIrIn₅ under pressure down to 0.3 K and the same conclusion has been reached.²⁷

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